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Applicant(s): Graylon K. Williams, et al

Docket No.

5702-00004

Serial No.

09/664,130

Filing Date

September 18, 2000

Examiner

Edward A. Miller

Group Art Unit

3641✓

Invention:

GAS GENERANTS CONTAINING SILICONE FUELS

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**GROUP 3800**

I hereby certify that the following correspondence:

Reply Brief and statement under 37 C.F.R. 1.195 (14 Pages) in Triplicate; Copy of Exhibit A (4 Pages); Copy of Exhibit B (4 Pages); Copy of Exhibit C (2 Pages); Check in the amount of \$420.00 for a Two-Month Extension of Time

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IN THE UNITED STATES PATENT  
AND TRADEMARK OFFICE

Group Art Unit 3641  
Examiner Edward A. Miller

Graylon K. WILLIAMS, et al.

REPLY BRIEF AND STATEMENT  
UNDER 37 C.F.R. 1.195

Serial No. 09/664,130

Filed September 18, 2000

For: GAS GENERANTS CONTAINING SILICONE FUELS/

November 3, 2003

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GROUP 3600

Sir:

Applicants presented a reply brief responsive to the Examiner's response on September 2, 2003. The Examiner refused to enter the brief stating that the brief had not been supplied in triplicate as required, and that the brief contained "evidence" not earlier submitted in contravention of 37 C.F.R. 1.193(b). See Paper No. 19. The undersigned has contacted Mr. Carone relative to Examiner Miller's concerns in Paper No. 19 and in response, Mr. Carone indicated that if a statement pursuant to 37 C.F.R. 1.195 (of why the "evidence" had not earlier been presented) is included with the reply brief, the reply brief would be entered. Applicants therefore resubmit the reply brief timely filed on September 2, 2003 and concurrently therewith, submit a statement under 37 C.F.R. 1.195 detailing why the exhibits submitted with the reply brief were not earlier presented. At the same time, pursuant to the examiner's comments in Paper No. 19, Applicants petition for a two-month extension for the period of response and include the appropriate fee.

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STATEMENT UNDER 37 C.F.R. 1.195

Applicants submit Exhibits A, B, and C responsive to comments first made by the examiner in the Examiner's Response to the Appeal Brief, having a mailing date of July 2, 2003. In the examiner's response on page 5, the examiner

responded to the applicants' argument that addition of carbon would increase the amount of carbon monoxide in contravention of Grebert (see applicants' argument on pages 4, 5, and 6 of the Appeal Brief). The applicants did not present the exhibits with the appeal brief because the understanding of how carbon monoxide is formed is so well known (see the Exhibits), that further support of the argument was not believed necessary. Specifically, the examiner stated:

"...As to carbonate coolant not being desirable in Grebert et al., due to carbon is not persuasive. Plantif et al. teach the use of various [alkali metal and alkaline earth metal] carbonate, oxalate, and bicarbonate coolants at col. 3, lines 23-26. In Hamilton, the coolant is preferably a carbonate, col. 2, lines 26-32. It must be observed that the carbon in a metal carbonate decomposes to form metal oxide and carbon dioxide. In other words, appellants' objection to adding carbon lacks scientific foundation, as the carbon of the coolant is already oxidized, or mostly so in the case of oxalate..."(emphasis added)

The examiner had not prior to the Examiner's Response Brief raised this argument and therefore the applicants only presented known textual material that would rebut the examiner's argument. The examiner's argument had not been earlier made and therefore the applicants reasonably and timely responded to rebut the examiner's argument in the Reply Brief. The exhibits do not consist of added technical data relative to the application but simply rebut the examiner's arguments quoted above. Denial of entry of the Reply Brief on the basis of inclusion of textual exhibits known in the art would deny the applicants a full prosecutorial opportunity to respond to arguments not earlier presented by the examiner.

Accordingly, the applicants courteously solicit entry of the reply brief with the exhibits given that a showing of good and sufficient reason for the earlier presentation of the exhibits has been shown.

#### REPLY BRIEF

Claims 1-13 and 16-18 stand rejected under 35 U.S.C. 103(a) as being unpatentable over Grebert et al. (3,986,908) in view of Plantif et al. (3,964,256), Hamilton (4,309,229), Ochi et al. (5,656,793), and Taylor et al. (5,538,568). Pursuant to 37 C.F.R. § 1.193(b)(1), Applicants now again file a reply brief in

response to the Examiner's Answer mailed July 2, 2003.

**Grebert et al.** teaches compositions containing metal and nonmetal perchlorates and silicone rubbers as organic binders. Grebert does not teach or suggest the use of coolants as presently claimed. Furthermore, as explained in the appeal brief (incorporated herein by reference), Grebert actually teaches away from the addition of other carbon-containing compounds.

**Plantif et al** recognizes the benefit of forming compositions containing perchlorate oxidizers and silicone resin. Plantif also recognizes the benefit of coolants, but only as coolant stages or phases separate from the combustion stage. Also note that Plantif describes the use of the coolants at temperatures well below the combustion temperature. The second coolant stage is affected by combustion gas contact with a solid particulate coolant comprising a compound which decomposes at a temperature below 200°C to produce a gas. Suitable compounds include alkali metal or alkaline earth metal carbonates, oxalates, or bicarbonates. See column 3, lines 20-35. As such, Plantif incorporates outboard coolants as recognized by the Examiner, but does not contemplate or suggest the use of a coolant integral to the gas generant composition that is then combusted at much higher temperatures. Accordingly, even though cooling the combustion gases is recognized as a problem, there simply is no suggestion by Plantif to integrate the coolant into the gas generant composition to accomplish cooling of the combustion gases. Therefore, when reviewing Plantif one of ordinary skill in the art would not be motivated to completely change the invention of Plantif to add the coolants into the gas generant composition rather than cool in stages subsequent to the combustion stage.

**Hamilton teaches away** from the present invention. At column 1, lines 45-50, Hamilton summarizes the invention:

In accordance with the present invention, a combustible mixture of carbon, an oxidizer which does not contain hydrogen and, optionally, a coolant are mixed to form a combustible material which produces non-toxic gas rapidly, at relatively low temperature, and

***without the production of water vapor.*** (Emphasis added)

Silicone, as described in Formulas 1 and 2 of the present invention, for example, often contains an abundant amount of hydrogen. This contravenes Hamilton because of the propensity for compositions containing silicone to produce at least minimal amounts of water. In fact, **Grebert** at column 5, lines 26-40, describes the same. Accordingly, when a reference teaches away from an invention it cannot suggest the same. Stated another way, one of ordinary skill in the art would not be motivated to include silicone in a gas generant composition in light of Hamilton because Hamilton specifically teaches against compositions that result in the formation of water vapor.

**Ochi et al.** does not suggest any fuel (or reducing agent) other than HDCA or hydrazodicarbonamide. The structure of HDCA is markedly different from silicone. Hence Ochi does not suggest the use of silicone by virtue of the use of HDCA. Compare Formula 1 of Ochi with Formulas 1 and 2 of the present invention. The examiner's attention is also directed to Tables 2, 5, and 6 as found in Ochi. The "In-tank gas temperature (°C)" resulting from combustion of compositions described by Ochi ranges from 152 to 407 °C. When viewed in light of Ochi's objects of the invention as described in column 2, lines 5-35 (e.g. low combustion temperature), one of ordinary skill in the art would not be motivated to replace the low heat generating HDCA with silicone. Nor would one of ordinary skill in the art be motivated to add silicone to the compositions of Ochi. Adding silicone would increase the temperature in contravention of the stated purpose of maintaining low temperatures. The use of silicone results in temperatures in excess of 2000°C as given in Table 1 on pages 6 and 7 of the specification. Therefore, Ochi et al. also **teaches away** from the present invention.

Although **Taylor** describes the use of perchlorates in compositions, the similarity ends there. As with the other cited references, Taylor does not offer any motivation or suggest formulation of the present compositions when viewed in light of Grebert. Taylor has as an object of the invention production of an easily

extrudable composition that is readily cured at room temperature, and mixed at low viscosity. See the "Summary of the Invention". These objects simply differ from the benefits characterized in the present invention. As such, one of ordinary skill in the art would not be motivated to consider Taylor when formulating the present compositions. A prima facie case of obviousness cannot be supported without the requisite showing of a suggestion or motivation to combine the references. Again, as with the other references mentioned above, Taylor falls short in this regard.

In view of the above, Applicants note that it is improper to combine references where the references teach away from their combination. *In re Grasselli*, 713 F.2d 731, 743, 218 USPQ 769, 779 (Fed. Cir. 1983) (The claimed catalyst which contained both iron and an alkali metal was not suggested by the combination of a reference which taught the interchangeability of antimony and alkali metal with the same beneficial result, combined with a reference excluding antimony from, and adding iron to, a catalyst.)

### ***Response to Examiner's Arguments***

In addition to the comments above, Applicants note the following relative to the Examiner's response to the Applicants' arguments presented in the appeal brief.

To begin with, the Examiner throughout refers to the variation of the specific type and amount of coolant as well as "other notoriously well known ingredients, for example, within the parameters taught, to obtain a suitable cooling result, would have been obvious to the person of ordinary skill in the art." See page 4, lines 5-6 of the Examiner's Response. In the same way, the Examiner again refers to coolants as "a notoriously well known variable that one of ordinary skill in the art may apply as desired". See page 6, lines 12-13 of the Examiner's Response. Other occurrences of the same or similar language are also noted. For

example, on page 6, lines 16-17, "all as is notoriously well known to one of ordinary skill in the art."

The Examiner is respectfully advised that, "a statement that modifications of the prior art to meet the claimed invention '*would have been well within the ordinary skill of the art at the time the invention was made*' because the references relied upon teach that all aspects of the claimed invention were individually known in the art is not sufficient to establish a prima facie case of obviousness without some objective reason to combine the teaching of the references." *Ex parte Levengood* 28 USPQ 2d 1300 (Bd. Pat. App. & Inter. 1993) See also *In re Kotzab* 217 F. 3d 1365, 1371, 55 USPQ 2d 1313, 1318 (Fed. Cir. 2000)

Furthermore, the Examiner is also advised that, "the question under 35 U.S.C. § 103 is not whether the differences [between the claimed invention and the prior art] would have been obvious" but "whether the claimed invention *as a whole* would have been obvious." *Stratoflex, Inc. v. Aeroquip Corp.*, 713 F.2d 1530, 1537, 218 USPQ 871, 877 (Fed. Cir. 1983) Accordingly, Applicants respectfully note that the Examiner's emphasis on addition and variation of the coolant is misplaced. Rather, proper emphasis is placed on the compositions as a whole and how the references taken as a whole suggest to one of ordinary skill in the art their respective combination to form the compositions as claimed in the present application.

In the same way, see page 6, lines 17-18 of the Examiner's Response, "Further, although in some instances as in Hamilton, water vapor may not be desired, this is also an art recognized choice to the ordinarily skilled artisan." The case law cited above is also pertinent here. Furthermore, the Examiner fails to appreciate that Hamilton must be viewed *as a whole* as should any other reference when applied under 35 U.S.C. 103. When viewed as a whole, it becomes apparent that Hamilton would not lead one of ordinary skill to combine the teachings of Hamilton with Grebert, for as discussed herein and in the Appeal Brief, each cited reference explicitly and/or implicitly teaches away from the

combination with the other. The Examiner states on page 4, lines 16-17 that "it is respectfully pointed out that it is the teachings of the prior art that are combined for what they teach, not the prior art per se." Applicants respectfully point out that it is the prior art, not the individual teachings, that provide the basis for the rejection under 35 U.S.C. 103. Accordingly, the Examiner is advised that the prior art references must be considered in their entirety, i.e., as a whole, including portions that would lead away from the claimed invention. *W.L. Gore & Associates, Inc. v. Garlock, Inc.*, 721 F.2d 1540, 220 USPQ 303 (Fed. Cir. 1983), *cert. denied*, 469 U.S. 851 (1984)

Accordingly, Applicants respectfully maintain that when the references are considered in their entirety rather than as individual teachings in the art as apparently urged by the Examiner, the impropriety of combining the references becomes apparent for the reasons stated by the Applicants in the Appeal Brief and throughout this response. Furthermore, Applicants maintain that the compositions incorporating coolants as claimed do not represent "notoriously well known" variables or constituents known for their use in silicone based compositions, or in gas generators and combusted therein. The cited references certainly would not lead to that conclusion. Further references are still requested. Applicants would direct the Examiner's attention to the arguments based on insufficient motivation to combine the cited references and also on how the various references actually teach away from their respective combination, particularly Grebert et al.

As to the arguments presented regarding the "comprising" scope, any distinctions made relative to the compositions described in the various references are not made to suggest what is not included in the present claims, but rather to emphasize what is not taught or motivated in the respective reference. Applicants appreciate the Examiner's remarks relative to the "comprising" scope in the claims and understand the term to be broad-based relative to the scope. Applicants also understand the term to mean that the named elements are essential, but other elements may be added and still form a construct within the scope of the claim. *Moleculon Research Corp. v. CBS, Inc.*, 793 F.2d 1261, 229 USPQ 805 (Fed. Cir.



1986); *In re Baxter*, 656 F.2d 679, 686, 210 USPQ 795, 803 (CCPA 1981)

**Grebert et al. – Insufficient Motivation to Combine**

As to the argument at the top of the Examiner's Response on page 5, Grebert et al. does teach that the gases should be non-toxic. Grebert also teaches that the compositions described must exhibit enhanced mechanical properties or mechanical strength. Accordingly, Grebert's compositions are directed at propellant compositions that have good mechanical strength and that yield combustion gases which are substantially free of toxic gases and which are, therefore, suitable for use as gas generators for inflatable cushion protection devices. See Grebert at column 1, lines 22- 35. As recognized by the Examiner, Applicants agree that non-toxicity is of course a concern with all gas generants involving human exposure. Nevertheless, in contradistinction to Grebert et al., the problems addressed by the present invention are centered about providing a gas generant composition having a sustained burn at ambient pressure while exhibiting relatively reduced temperatures. See the Background of the Invention and see Table 1 and the Examples in the present application. Because Grebert et al. describes a different purpose for different compositions, Applicants maintain that one of ordinary skill in the art would not be motivated to combine Grebert et al. with Plantif, Hamilton, Taylor, or Ochi, because as explained above, the purposes for Grebert depart from the purposes of the present invention. Stated another way, Applicants are primarily concerned with sustained combustion at ambient pressures with relatively lower temperatures. On the other hand, Grebert is primarily concerned with non-toxicity and higher mechanical strength.

In support thereof, Applicants respectfully note that, "there are three possible sources for a motivation to combine references: the nature of the problem to be solved, the teachings of the prior art, and the knowledge of persons of ordinary skill in the art." *In re Rouffet*, 149 F.3d 1350, 1357, 47 USPQ2d 1453, 1457-58 (Fed. Cir. 1998) (The combination of the references taught every

element of the claimed invention, however, without a motivation to combine, a rejection based on a *prima facie* case of obvious was held improper.) The level of skill in the art cannot be relied upon to provide the suggestion to combine references. *Al-Site Corp. v. VSI Int'l Inc.*, 174 F.3d 1308, 50 USPQ2d 1161 (Fed. Cir. 1999) Applicants note that the Examiner has not shown the requisite motivation or suggestion to combine Grebert et al. with the other cited references as required to support a *prima facie* case of obviousness.

#### **Grebert et al. – Teaching Away**

With regard to **Grebert et al.** teaching away from combination with any other carbon-containing constituents, Applicants reiterate the discussion found in the Appeal Brief, herein incorporated by reference. Furthermore, Applicants acknowledge the Examiner's argument that the carbon in a metal carbonate decomposes to form metal oxide and carbon dioxide. The Examiner further states that appellants' objection to adding carbon lacks scientific foundation as the carbon of the coolant is already oxidized, or mostly so in the case of oxalate. Responsive thereto, Applicants respectfully maintain that it is well known in the art that carbon dioxide and carbon monoxide exist in equilibrium across a broad spectrum of pressures and temperatures. As such, addition of a coolant that decomposes to carbon dioxide (among other things) and would result in increased amounts of carbon monoxide in the combustion products. In support thereof, Applicants direct the Examiner's attention to **Exhibits A, B, and C**.

**Exhibit A** is a copy of an excerpt from *INORGANIC REACTIONS AND STRUCTURE*, Edwin S. Gould (1955, 1962) Holt, Rinehart and Winston, Inc. As shown on page 107, the decomposition of various alkaline earth carbonates occurs at different respective temperatures, or, the decomposition temperature of a carbonate differs with the alkaline earth metal (or metal) attached thereto.

**Exhibit B** is a copy of an excerpt from *Chemistry of the Elements*, N.N. Greenwood and A. Earnshaw (1984) Pergamon Press Ltd. As described in the

highlighted portions originally taken from the J. Inorg. Nucl. Chem. 1769-71 (1981),

"Very recently it has been shown that, at all pressures, there is a fairly wide range of temperatures in which CO<sub>2</sub> dissociates directly into CO and O<sub>2</sub> without precipitation of carbon.



As shown, CO and CO<sub>2</sub> exist in equilibrium in a broad variety of pressure/temperature permutations.

Exhibit C is a copy of an excerpt from J. Iron Steel Inst. , F.D. Richardson and J.H.E. Jeffes 160, 261 (1948). Note that,

"The equilibrium concentration of CO is 10% at 550°C and 99% at 1000°C.."

As shown, CO exists in equilibrium in greater percentages at higher temperatures.

It can therefore be concluded that at the temperatures indicated in Table 1, well above 1000°C, equilibrium conditions at the temperatures indicated would favor the formation of carbon monoxide. Accordingly, it can be concluded that the addition of metal carbonates and/or oxalates that result in carbon dioxide also result in a certain equilibrium concentration of carbon monoxide depending on the attendant temperature and pressure.

As such, the Examiner's assertion that the Appellants' position (*that the addition of a coolant such as a carbonate or oxalate adds carbon monoxide to the combustion gases*) is scientifically unfounded is respectfully traversed, especially in view of the textual references provided. Accordingly, Applicants again maintain that Grebert et al. teaches away from adding compounds or constituents that would increase the carbon monoxide because of the equilibrium conditions shown.

As to other arguments presented by the Examiner, Applicants note the following:

1. Contrary to the Examiner's assertion, Plantif et al. does not teach that the SNPE silicone gas generating composition may generate excess oxygen at column 2, lines 23-27. For the sake of argument only, even if the Examiner had shown this teaching to be the case, and even if the suggestion to combine Plantif with Grebert had been shown, Grebert describes the addition of aluminum. It is well known that aluminum scavenges the available oxygen to form aluminates thereby reducing the availability of oxygen for complete combustion of carbon monoxide. Again, this is illustrated in the table found in Grebert in column 4, lines 37-45.
2. The Examiner asserts that *"the problem Grebert et al. discusses is, obviously a translation difficulty from the French, is not any additive that is carbon containing, but an additive that is contained in the composition and is elemental carbon."* The Examiner's attention is directed to Grebert et al., column 4 line 14-column 5 line 15. In contrast to what the Examiner urges, note that Grebert teaches that the addition of a binder should be reduced to reduce the production of carbon monoxide. He does not focus on elemental carbon but on carbon-containing binders (e.g. silicone). Applicants note that this same admonition would be applicable to any carbon-containing compound likely to produce carbon monoxide. The notion that this is a translational error would require that the same translational difficulty occur throughout the reference. Applicants still do not understand how the Examiner concludes that the problem is limited to elemental carbon. It plainly is not, **as taught by Grebert et al.**
3. The Examiner alludes to the breadth of the present claims with

regard to the small amounts of aluminum fuel and carbon combustion catalysts. Applicants respectfully maintain that the proper focus is placed on the motivation of one of ordinary skill in the art to combine the cited references. It is only placed on the claims with regard to the essential limitations described therein. It is unclear why the Examiner focuses on the breadth of the claims instead of on the motivation of one of ordinary skill in the art to combine the references to form a prima facie case of obviousness. The Examiner has not shown why one of ordinary skill in the art would combine the references to formulate the essential elements of the claims as presently given. In essence, a prima facie case of obviousness is not predicated on the breadth of the claims and what they might include. Rather, a prima facie case of obviousness is supported by references *properly combined* that teach or describe all of the essential limitations of the claims. Accordingly, Applicants continue to maintain that for the reasons stated, a prima facie case of obviousness is simply not supported.

4. Applicants note the Examiner's instruction relative to Ochi and various facts about coolants: *"These are not opinion, they are facts. Coolants cool endothermically; that is a matter of plain chemistry."* Applicants agree. See page 2, lines 23-26 of the Appeal Brief, for example. Also, the Examiner states, *"Among the variables are the metals and construction of the gas generating apparatus, as well as the presence or absence and the length of ducts, and variation in the materials of the air bag itself."* Applicants respectfully note that these "variables" are not implicated in the claims. Rather, unique compositions incorporating a coolant are claimed by the Applicants. These are not notoriously well known or Plantif et al, for example, would certainly have alluded to their use within a composition rather than to the use of outboard coolants as taught therein.

5. With regard to Ochi et al., Applicants note that Ochi describes the use of coolants, but not within compositions containing silicone and perchlorate oxidizers. In fact, none of the references describe coolants used within a composition containing silicone and a perchlorate oxidizer. Furthermore, because of different purposes or because of teaching away, the Examiner has not shown any motivation to combine the references. The Examiner is reminded that, "a statement that modifications of the prior art to meet the claimed invention '*would have been well within the ordinary skill of the art at the time the invention was made*' because the references relied upon teach that all aspects of the claimed invention were individually known in the art is not sufficient to establish a prima facie case of obviousness without some objective reason to combine the teaching of the references." *Ex parte Levengood* 28 USPQ 2d 1300 (Bd. Pat. App. & Inter. 1993) See also *In re Kotzab* 217 F. 3d 1365, 1371, 55 USPQ 2d 1313, 1318 (Fed. Cir. 2000)

Therefore, since none of the prior art of record teaches Applicants' claimed invention, nor provides any reason or motivation to modify the prior art to solve the specific problems for which Applicants were concerned, Applicants' position is that the Examiner has improperly relied upon Applicants' own teachings to support the grounds of rejection. See *In re Regel*, 526 F. 2d 1399, 1403 n.6, 188 USPQ 136, 139 n. 6 (CCPA 1975) ("there must be some logical reason apparent from positive, concrete evidence of record which justifies a combination of primary and secondary references") (citing *In re Stemniski*, 444 F.2d 581, 170 USPQ 343 (CCPA 1971)); *In re Geiger*, 815 F.2d 686, 688 2 USPQ2d 1276, 1278 (Fed. Cir. 1987) (obviousness cannot be established by combining pieces of prior art absent some "teaching, suggestion, or incentive supporting the combination"); *In re Cho*, 813 F.2d 378, 382, 1 USPQ2d 1662, 1664 (Fed. Cir. 1987) (discussing

the Board's holding that "the artisan would have been motivated" to combine the references); *In re Deminski*, 796 F.2d 436, 443, 230 USPQ 313, 316 (Fed. Cir. 1986) (impropriety of hindsight reconstruction); and *In re Donohue*, 766 F.2d 531, 534, 226 USPQ 619, 622 (Fed. Cir. 1985) (referring to the "suggestion or motivation to combine teachings" in rejections for obviousness) (citing *In re Samour*, 571 F.2d 559, 563, 197 USPQ 1, 4-5 (CCPA 1978)). As such, the Examiner has failed to properly set forth a prima facie case of obviousness.

In sum, none of the references when taken alone or when taken together suggest or describe the present invention. Stated another way, for the reasons given, a prima facie case of obviousness cannot be supported by references that teach away from the present invention, nor can it be supported without the requisite showing of a motivation to combine the references.

Accordingly, applicants respectfully traverse the rejection of claims 1-13 and 16-18 and courteously solicit the allowance of these claims and passage of the subject application to issue.

Applicants have calculated a fee of \$420.00 to be due in connection with this paper and our check in that amount is enclosed. The Commissioner is hereby authorized to charge Account No. 04-1131 for any deficiency or credit any overage to the same account. A duplicate copy of the first page of this transmittal is also included.

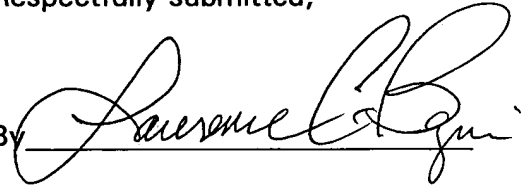
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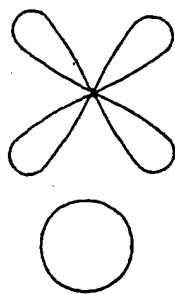
By



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## Exhibit A





# INORGANIC REACTIONS AND STRUCTURE

Revised Edition

**EDWIN S. GOULD**  
Stanford Research Institute  
(Formerly Polytechnic Institute of Brooklyn)

HOLT, RINEHART AND WINSTON · NEW YORK

## PREFACE TO REVISED EDITION

In the revision of this text, the plan of presentation used in the first edition is, in essence, retained. Major changes, however, have been made in discussions of a number of areas in which recent progress has been most striking. In line with the tremendous increase in interest in inorganic reaction mechanisms, a chapter has been added on that topic; and sections have been added describing electron spin resonance and nuclear magnetic resonance spectroscopy. In discussions of complexes of the transition metals I have used, wherever possible, the ligand-field approach rather than the less powerful valence-bond approach (which, until the early 1950's, was an important part of the structural thinking of inorganic chemists).

As in the first edition, many of the descriptions are brief and non-mathematical in nature. However, the supplementary reading lists at the ends of the chapters, which have been expanded and brought more nearly up to date, contain references to more advanced and rigorous treatments. Some exercises have been changed, some added, a few deleted, and asterisks have been used to mark those which experience has shown to be most challenging to the more capable students. Teachers may obtain answer sheets for exercises in this edition by writing the publisher.

It is a pleasure to thank the members of the Inorganic Chemistry Division at the Polytechnic Institute of Brooklyn and many former students, both graduate and undergraduate, for drawing my attention to errors, both large and small, that appeared in the first edition. I am very much indebted to the Physical and Inorganic Chemistry Section at Stanford Research Institute for support during this revision. My thanks go also to Miss Corlee Lukow for help in preparing the revised manuscript and, once again, to my wife Marjorie for help in countless details.

February, 1962

E. S. G.

Menlo Park, California

TO PROFESSOR LINUS PAULING,

*one of my early instructors in college chemistry,  
who also finds time to do research*

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contact with water for days at room temperature. The alkaline-earth metals differ also from the alkali metals in that the lighter members of the divalent group can form some complex ions. Although formation of complex ions of the alkali metals is not completely unknown, it is quite rationally limited, in general to aquo-complex formation.

### PREPARATIONS

For many years electrolysis of the fused halides was virtually the only method for obtaining these metals in free state, and it is still very much the method of choice for preparing such metals in small quantity. Recently a number of high temperature reductions have assumed importance; methods using aluminum for reduction of CaO, coal for the reduction of MgO, ferrosilicon alloy for the reduction of MgO, and silicon for reduction of BaO have been developed.

### TRENDS WITHIN THE SERIES

Much that has been noted concerning the trends within the alkaline metal family can be applied also to the alkaline earths. As the radius of the atom increases, removal of the outer electrons becomes easier; both the ionization potentials for the free atoms and the  $E^\circ$  values for the metals show this:

Be: radius 0.9 Å; ionization potential 9.3 volts;  $E^\circ$  1.7 volts  
Ba: radius 2.0 Å; ionization potential 5.2 volts;  $E^\circ$  2.9 volts

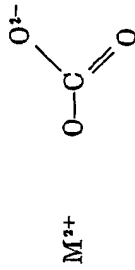
The values for magnesium, calcium, and strontium lie between those known for beryllium and for barium. All the trends are continuous; there is no anomaly due to hydration energies as was illustrated for lithium. It should be noted once more that ionization potentials and  $E^\circ$  values commonly listed describe reducibility from opposite viewpoints; a very good reducing agent will have a low value for its  $IP$  but a high  $E^\circ$  value.) The second ionization potentials of the alkaline-earth metals—at least, the energies required to remove the second valence electrons—are about twice as great as the first  $IP$ 's. (For Be,  $IP_1 = 9.3$  volts and  $IP_2 = 12$  volts. For Ba,  $IP_1 = 5.2$  volts and  $IP_2 = 10.0$  volts.) Such figures indicate that removal of the second electron is about twice as difficult as removal of the first and suggest that these metals should be univalent rather than divalent. However, the heat of hydration for a divalent ion is so much greater than for a univalent ion (Mg, 460 kcal; Na, 97 kcal) that the extra energy released in hydration of the divalent ions more than compensates for the extra energy necessary to remove the second valence electrons.

As with the other families, the oxides and salts of the lighter and

smaller-sized members are more covalent than those of the heavier and larger-sized members. The contrast between the amphoteric oxide of beryllium and the very basic oxide of barium should be familiar. Let us consider also a trend in the stabilities of the alkaline-earth carbonates, in particular, the temperatures needed to decompose these carbonates at one atmosphere pressure of carbon dioxide:

BaCO <sub>3</sub>	SrCO <sub>3</sub>	CaCO <sub>3</sub>	MgCO <sub>3</sub>	BeCO <sub>3</sub>
1360°	1290°	900°	540°	less than 100°

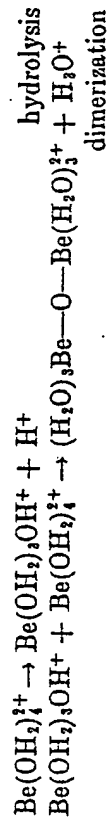
The crystal structures of these solid carbonates are such that it cannot be said that one particular metal ion "belongs" to one particular carbonate ion; nevertheless, a portion of the structure may be represented schematically:



In the most stable carbonates, the interaction between  $M^{2+}$  and the nearby oxygen atom of the  $\text{CO}_3^{2-}$  ion is purely electrostatic; there is comparatively little electron density between the two atoms. As the density of positive charge on  $M^{2+}$  increases, the electron cloud on the nearby oxygen becomes distorted toward  $M^{2+}$ , the adjoining O—C bond becomes weakened, and the O—M bond becomes strengthened, favoring breakup of the carbonate to the oxide and  $\text{CO}_2$ . If the positive ion has a very great positive-charge density ( $\text{Al}^{3+}$ ,  $\text{H}^+$ ), the carbonate cannot be made at all.

### THE ANOMALY OF BERYLLIUM

As with lithium, many properties of beryllium stand distinctly apart from those of its congeners. Again, the tiny size of the beryllium ion is responsible for the peculiarities. If we arbitrarily call the volume of the magnesium ion 1.0 unit, the volumes of the calcium, strontium, and barium ions become, respectively, about 3, 5, and 8 units; however, the volume of the beryllium ion is, on the same scale, only 1/8 unit. Since the oxide is amphoteric, one would expect many of the salts of  $\text{Be}^{2+}$  to be extensively hydrolyzed in water (as is the case with the salts of  $\text{Al}^{3+}$ ,  $\text{Zn}^{2+}$ , and  $\text{Cr}^{3+}$ ). The acidity of beryllium-containing solutions is also increased by polymerization of the beryllium-containing ions:



Beryllium far surpasses its congeners in its ability to form complexes. Besides the stable  $\text{BeF}_4^{2-}$  ion, beryllium forms many complexes having

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Exhibit B

# **Chemistry** **of the** **Elements**

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## Forewo

THE publication of a new event of major importance: textbooks of inorganic chemistry and do not give reactions of the elements fills a real need for an update of elements. The facts concerning the essence of chemistry. Facts and theories. Moreover, it is a background of facts in chemistry and their usefulness and the random will quickly reveal unexplained and which theories. The writing of which is congratulated on having elements in such a real inorganic chemistry for every success.

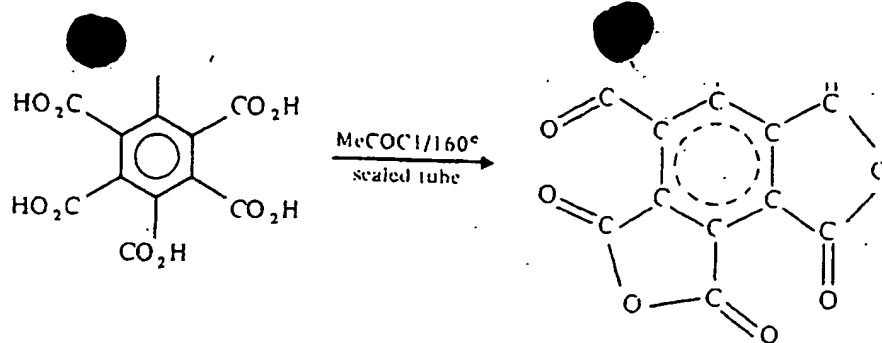


FIG. 8.14

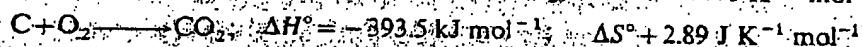
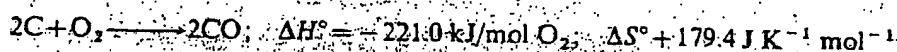
C-O bond confers considerable thermal stability on these molecules but the compounds are also quite reactive chemically, and many of the reactions are of major industrial importance. Some of these are discussed more fully in the Panel.

The nature of the bonding, particularly in CO, has excited much attention because of the unusual coordination number (1) and oxidation state (+2) of carbon: it is discussed on p. 349 in connection with the formation of metal-carbonyl complexes.

Pure CO can be made on a laboratory scale by dehydrating formic acid (HCOOH) with conc  $\text{H}_2\text{SO}_4$  at  $\sim 140^\circ$ . CO is a colourless, odourless, flammable gas; it has a relatively high toxicity due to its ability to form a complex with haemoglobin that is some 300 times

### Industrially Important Reactions of Oxygen and Oxides with Carbon

Carbon monoxide is widely used as a fuel in the form of producer gas or water gas and is also formed during the isolation of many metals from their oxides by reduction with coke. Producer gas is obtained by blowing air through incandescent coke and consists of about 25% CO, 4%  $\text{CO}_2$ , and 70%  $\text{N}_2$ , together with traces of  $\text{H}_2$ ,  $\text{CH}_4$ , and  $\text{O}_2$ . The reactions occurring during production are:



Water gas is made by blowing steam through incandescent coke. It consists of about 50%  $\text{H}_2$ , 40% CO, 5%  $\text{CO}_2$ , and 5%  $\text{N}_2 + \text{CH}_4$ . The oxidation of C by  $\text{H}_2\text{O}$  is strongly endothermic:



Consequently, the coke cools down and the steam must be intermittently replaced by a flow of air to reheat the coke.

At high temperatures, particularly in the presence of metal catalysts, CO undergoes reversible disproportionation†



† Very recently it has been shown that, at all pressures, there is a fairly wide range of temperatures in which  $\text{CO}_2$  dissociates directly into CO and  $\text{O}_2$  without precipitation of carbon.



For example, the temperature range is  $250\text{--}370^\circ\text{C}$  at  $10^{-2}$  atm,  $320\text{--}480^\circ\text{C}$  at 1 atm, and  $405\text{--}630^\circ\text{C}$  at 100 atm. At higher temperatures in each case, C is also formed, but always in the presence of some  $\text{O}_2$ .

<sup>11a</sup> M. H. LIETZKE and C. MULLINS, Thermal decomposition of carbon dioxide, *J. Inorg. Nucl. Chem.* 43, 1769–71 (1981).

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Exhibit C



The equilibrium concentration of CO is 10% at 550°C and 99% at 1000°C. As the forward reaction involves a reduction in the number of gaseous molecules in the system it is accompanied by a large decrease in entropy. Remembering that  $\Delta G = \Delta H - T\Delta S$  this implies that the reverse reaction becomes progressively more favoured at higher temperatures. The thermodynamic data for the formation of CO and CO<sub>2</sub> can be represented diagrammatically on an Ellingham diagram (Fig. A) which plots standard free energy changes per mol of O<sub>2</sub> as a function of the absolute temperature.

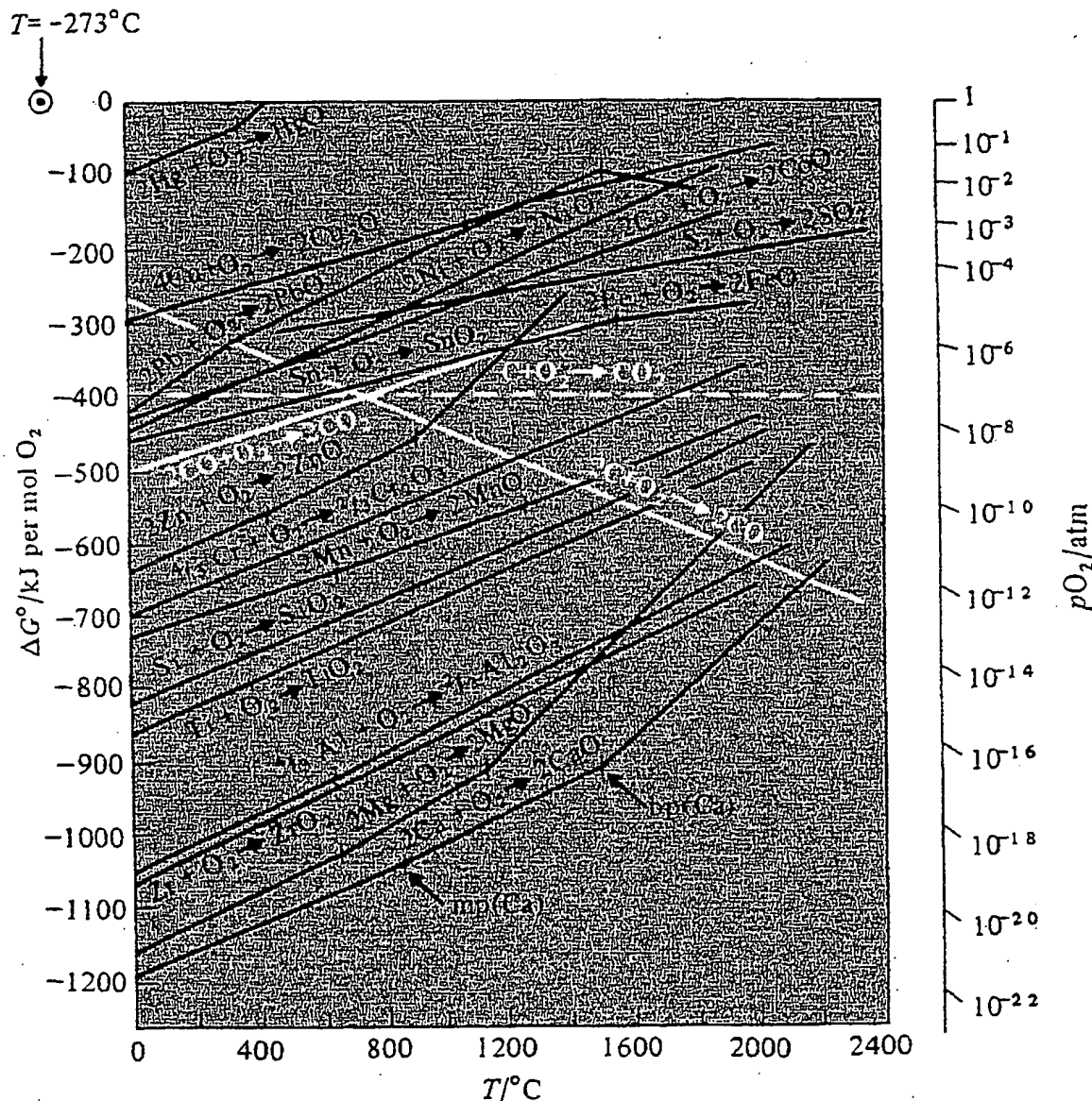


FIG. A Ellingham diagram for the free energy of formation of metallic oxides. (After F. D. Richardson and J. H. E. Jeffes, *J. Iron Steel Inst.* 160, 261 (1948).) The oxygen dissociation pressure of a given M-MO system at a given temperature is obtained by joining  $\odot$  on the top left hand to the appropriate point on the M-MO free-energy line, and extrapolating to the scale on the right hand ordinate for  $p_{O_2}$  (atm).

The oxidation of C to CO results in an increase in the number of gaseous molecules, it is therefore accompanied by a large increase in entropy and is favoured at high temperature. By contrast, oxidation to CO<sub>2</sub> leaves the number of gaseous molecules unchanged, there is little change in entropy ( $\Delta S^\circ 2.93 \text{ J K}^{-1} \text{ mol}^{-1}$ ), and the free energy is almost independent of temperature. The two lines (and that for the oxidation of CO to CO<sub>2</sub>) intersect at 983 K, it follows that  $\Delta G$  for the disproportionation reaction is zero at this temperature. The diagram also includes the plots of  $\Delta G$

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